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14. ABSTRACT  During the three years of this grant, conclusions were reached that are important for the future of semiconductor cavity QED utilizing photonic crystal nanocavities. A new way was found to scan the cavity mode by condensation of xenon or nitrogen while keeping the quantum dot cold to minimize dephasing. Absorption was shown to reduce the cavity Q when the dot density is 400 per square micron. Lasing with a soft threshold was observed using several quantum dots. Almost 30 samples were grown and characterized for the 900-1000 nm range; ten were grown with an AlGaAs sacrificial layer and GaAs slab for nanocavity fabrication at Caltech. Desirable characteristics were measured: peak wavelengths of the ensemble photoluminescence between 915 and 1020 nm, dot densities from 200 down to just a few per square micron, ensemble radiative lifetimes around 700 ps, and strong Purcell cavity enhanced emission exhibiting anti-bunched photon statistics. However, after at least five years of trying, only one of the best etching groups in the world achieved a Q exceeding 10,000 in the 900-1000 nm range using a GaAs slab; this strongly points to fabricating nanocavities for 1500 nm where scattering losses are smaller.					
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# **Deterministic Entangled Nanosource**

## **FINAL REPORT**

**August 2008**

**For September 1, 2005 through July 31, 2008**

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## Executive Summary

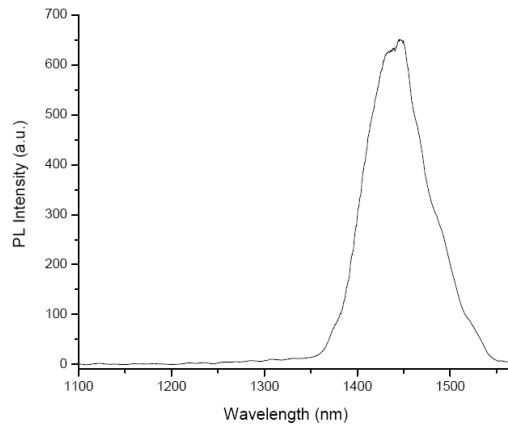
### Summary of the Most Important Results, Conclusions, and Future Directions

During the three years of this grant, conclusions have been reached that are important for the future of semiconductor cavity QED utilizing photonic crystal nanocavities. When this grant began, we expected that we would be able to get much higher Qs than the 6000 in our original publication in Nature at the end of 2004 [Yoshie et al. 2004]. Consequently we planned to purchase the best available detectors for 1200 nm and use them to study photon statistics. Even though we found cavities with Qs up to 20,000 on the low energy tail of the quantum dot (QD) ensemble distribution [Hendrickson et al. 2005a,b and 2006a,b], we did not find any SQDs giving strong coupling in spite of developing a new way of scanning the cavity mode by condensation of xenon or nitrogen while keeping the QD cold to minimize dephasing [Mosor et al. 2005]. We concluded that working with self-assembled InAs QDs at the long wavelength limit of their growth range might not be giving suitable dots for strong coupling. We speculated that by growing dots with a shorter wavelength ensemble peak, one could work on the long wavelength edge with standard dots or reduce the density and work near the peak. An additional advantage would be that Si detectors work much better at 900 nm than InGaAs detectors work at 1200 nm. We grew and characterized almost 30 QD samples for the 900-1000 nm range, ten of them with an AlGaAs sacrificial layer and GaAs slab designed for photonic crystal slab nanocavity fabrication. We grew very good QDs as judged by a) the peak wavelength of the ensemble PL (915-1020 nm), b) the density of surface dots (200 down to just a few per square micron by AFM), c) the ensemble radiative lifetime (700 ps by streak camera following ps excitation [Sweet et al. 2007]), d) strong Purcell cavity enhanced emission, and e) Hanbury Brown Twiss measurements of the second order correlation function of strongly enhanced SQDs excited nonresonantly both cw and pulsed [Richards et al. 2007; Gibbs & Khitrova 2007; Gibbs 2007 & 2008a,b; Khitrova et al. 2007; Khitrova 2008a,b,c]. The cavity fabrication was performed by Uday Khankhoje in the Caltech group of Axel Scherer, since Tomo Yoshie graduated and became an Assistant Professor at Duke shortly after our Nature article. Uday gradually came up to speed, hampered by building construction, new instrumentation, and equipment breakdowns. His cavity Q gradually increased up to as high as 9000 on our 900-1000 nm samples.

Several other groups continued their quest for strong coupling using photonic crystal nanocavities which have one big advantage: the smallest mode volume and hence the highest vacuum field and thus the largest vacuum Rabi splitting for the same dipole moment [Khitrova et al. 2006]. However, it was not until 2007 that first the group of Imamoğlu [Hennessy et al. 2007] and then the group of Vuckovic [Englund et al. 2007]) reported strong coupling. Both succeeded in seeing strong coupling at short wavelengths (925-950 nm) with almost the same splitting to linewidth ratio as our 2004 Nature article. Hennessy et al. fabricated a photonic crystal nanocavity around a good isolated SQD previously identified. Englund et al. demonstrated strong coupling in reflectivity rather than PL as we and others used before. These were important advances, but it is unlikely that we could have published either in Nature having published there the much more important first observation.

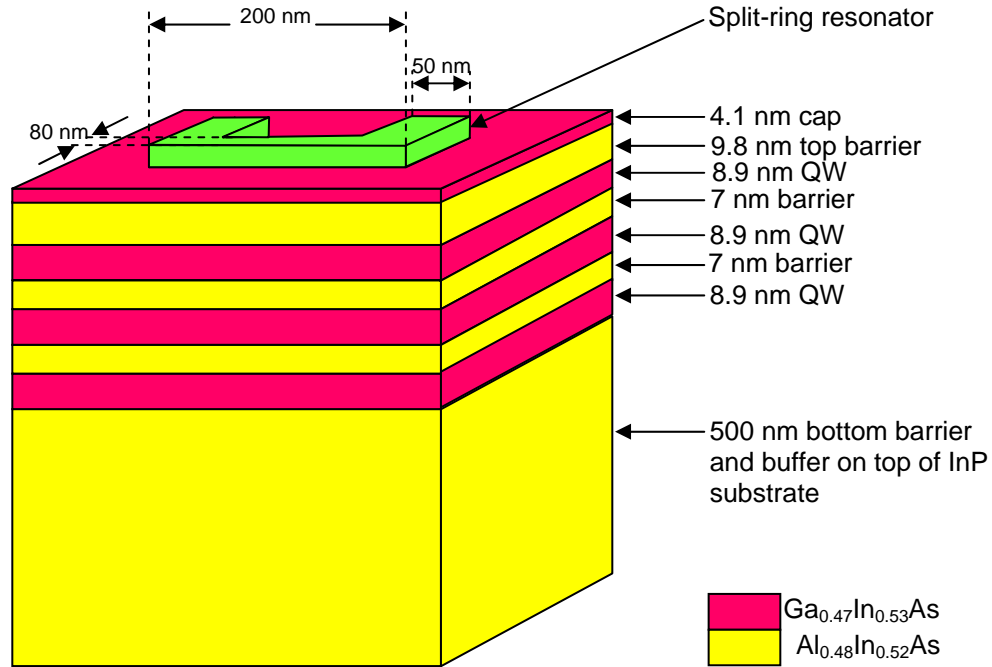
The highest Q reported by the group of Forchel, which has obtained Qs of 150,000 for micropillar cavities, is 9,000 [Sünner et al. 2008]. The highest Qs reported by the groups of Imamoğlu and Vuckovic have been under 10,000 in all but one case which reached 20,000 [Fushman et al. 2008]. Private communication suggests that the latter involved surface passivation. That Uday's fabrication technique approaches that of Tomo Yoshie is evidenced by his obtaining Qs as high as 25,000 at 1200 nm on the same MBE sample used for our Nature strong coupling. The fact that after at least five years of trying, only one of the best etching groups in the world has succeeded in obtaining a Q exceeding 10,000 at 915 nm using a GaAs slab strongly suggests that there is a fabrication limitation that is not likely to be overcome. Since scattering losses scale as the inverse fourth power of the wavelength, they are reduced by a factor of 3 by increasing the wavelength from 915 to 1200 nm. Moving from 1200 to 1500 nm would reduce them by another factor of 2.5. Rather than go back to 1200 nm which we investigated thoroughly, this reasoning points to going to 1500-1600 nm. This goal involves new challenges, namely growing suitable QDs and fabricating nanocavities with an  $\text{Al}_{0.48}\text{In}_{0.52}\text{As}$  sacrificial layer grown lattice matched on an InP substrate.

As a first step, we have grown InAs QDs that photoluminesce in the 1400-1500 nm range at low temperature (HSG4); see Fig.1. The growth parameters were guided by the results reported in [Enzmann et al. 2007].



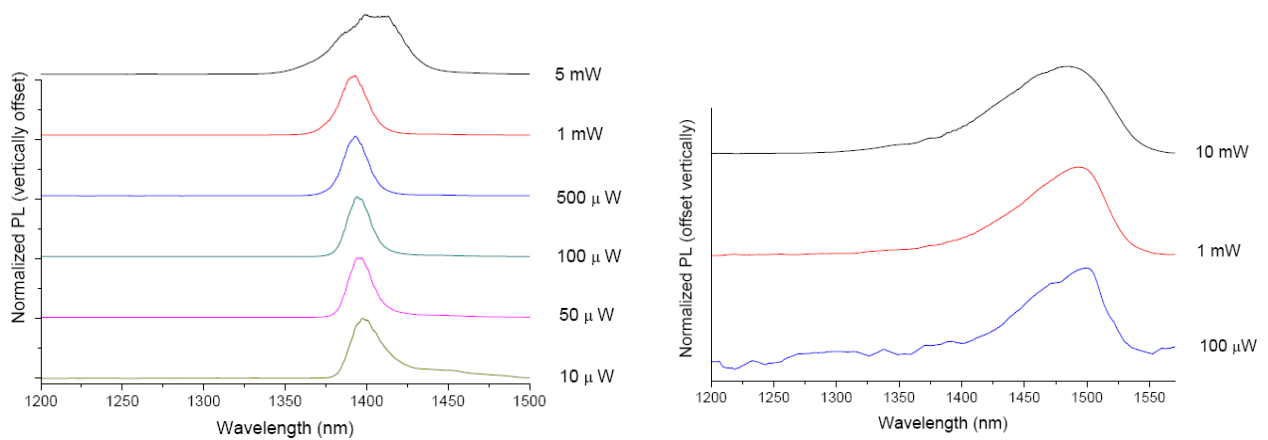
*Fig.1 Photoluminescence spectrum of an ensemble of QDs in a single layer excited cw weakly at 780 nm.*

Whereas the results of our experiments over the last three years are driving us toward 1500 nm for our strong coupling nanocavities, the group of Martin Wegener in Karlsruhe has independently concluded that 1500 nm is the shortest wavelength for which it is able to fabricate good split-ring resonators. Such metallic devices are inherently quite lossy, so that any applications will depend upon fabricating them on top of a material that can supply gain to compensate for these losses. We are collaborating with him to evaluate the use of quantum wells and quantum dots to provide this gain. Since the evanescent tail of the plasmonic resonance decays within 50 nm into the GaAs, the gain material must be grown close to the surface. In addition to the QDs above, we have grown two SQW samples and one with three QWs; the layer structure of the latter (HSG3) is shown in Fig.2.



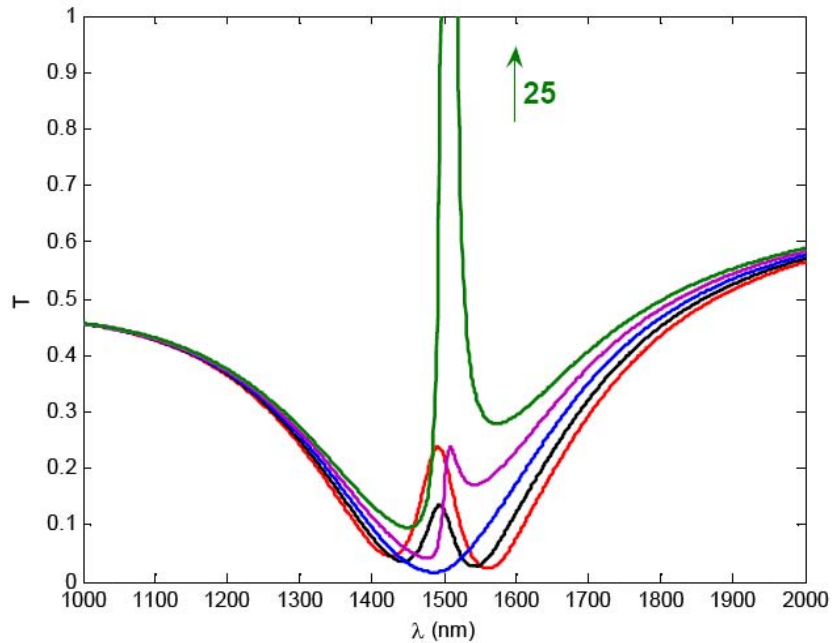
*Fig.2 Layer structure of sample HSG3 consisting of three GaInAs QWs between AlInAs barriers grown by MBE on an InP substrate.*

The PL spectra from the three-QW sample have been studied as a function of excitation power both at low and at room temperature; see Fig.3. The intensity of the PL increases linearly with increased pump power. No measurements of gain have been made yet.



*Fig.3 Normalized photoluminescence from three-QW sample HSG3 at (left) 10 K and at (right) 300 K. Note the good overlap with the 1500-nm target wavelength for split-ring resonator gain at room temperature.*

One of the major challenges in the emerging field of photonic metamaterials [Shalaev 2007; Soukoulis et al. 2007] lies in significantly reducing or compensating the losses. For example, the record-high figure of merit of  $FOM = 3$  of the negative-index metamaterial operating around  $1.4\text{-}\mu\text{m}$  wavelength reported in [Dolling et al. 2006] still translates into an effective absorption coefficient of  $\alpha = 3 \times 10^4 \text{ cm}^{-1}$  – which is even larger than the band-to-band absorption of typical direct-gap semiconductors such as, e.g. GaAs (there,  $\alpha \approx 10^4 \text{ cm}^{-1}$ ). Thus, at first sight, it seems hopeless to compensate that level of absorption by gain. Yet, an interesting recent theoretical publication [Zheludev et al. 2008] shows that it is *not* the bulk gain coefficient that matters but rather the *effective* gain coefficient of the combined system. Due to pronounced *local-field* enhancement effects in the spatial vicinity of the metallic nanostructure (e.g. the split-ring resonator illustrated in Fig.2, the effective gain coefficient can be substantially larger than its bulk counterpart). A gold (Au) split-ring resonator of the dimensions given in Fig.2 has an optical resonance close to  $1500 \text{ nm}$ . This plasmonic oscillator is able to couple to other oscillators such as a semiconductor resonance. To better understand the interaction of a plasmonic and semiconductor resonance, one can employ a toy model in which the latter is approximated by a fermionic two-level system and the plasmonic resonance by a bosonic resonance [Wegener et al. 2008]. Some numerical results from the toy model are displayed in Fig.4.



*Fig.4 Steady-state transmittance  $T$  versus wavelength  $\lambda$  of a structure similar to Fig.2 according to the closed analytical solution of a simple toy model for fixed two-level-system occupation  $f = 0$  (red),  $0.25$  (black),  $0.5$  (blue),  $0.75$  (magenta), and  $1$  (green). The model parameters have been chosen to roughly match those of a typical array of split-ring resonators located on a  $50\text{-nm}$  thin film composed of semiconductor quantum dots. An additional background dielectric constant of  $13.8$  accounts for the semiconductor gain film (e.g. InGaAs).*

Figure 4 shows the evolution of the transmittance spectra for increasing two-level-system upper-state occupation ( $f = 0, 0.25, 0.50, 0.75$ , and  $1$ ). Notably, the line width of the sharp transmittance maximum in Fig.4 goes to zero as the transmittance peak approaches and eventually exceeds unity. At this point, the gain effectively compensates the loss. Hence, we interpret this feature as being indicative for the onset of lasing (or “spasing” [Bergman & Stockman 2003] or “lasing spasing” [Zheludev et al. 2008]). Note that the transmittance shoots out of the frame in Fig.4 and becomes  $T = 25$  for  $f = 1$ . This simplified figure assumes that the 2LS linewidth is independent of  $f$ .

The PI's senior graduate student, Josh Hendrickson, after attending NOEKS9 in Klink (close to Berlin), visited the labs of Martin Wegener in Karlsruhe. Josh is spending September there working with Dr. Stephan Linden (Institut für Nanotechnologie) to learn how to sputter on silica, spin on photoresist, e-beam write, and remove developed resist in order to produce silica masks for MBE growth of quantum dots in Tucson. He will also help in any way he can with the fabrication of split-ring resonators on our semiconductor gain samples and their testing via femtosecond pump/probe spectroscopy. Josh will concentrate on this project until he completes his Ph.D. next May; then he has accepted an offer to be a postdoc working jointly with Martin Wegener and the PI.

During this period we grew 18 samples to study radiative coupling effects between QWs spaced, not periodically as usual, but instead nonperiodically with Fibonacci spacings. This structure still has a photonic stopband, but it is impossible for all of the QWs to be at nodes of the optical field as they are in the periodic case. Consequently, this Fibonacci 1D quasicrystal emits strong photoluminescence normal to the sample when the Bragg condition is satisfied. In contrast, a periodic 1D crystal has almost no photoluminescence under this condition. Two talks [Khitrova 2008b,c] were presented and an Optics Express [Hendrickson et al. 2008] was accepted on these nonperiodic structures.

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- Gibbs, H. M., and G. Khitrova, "Strong coupling between an InAs single quantum dot and a photonic crystal slab nanocavity," University of Würzburg, Germany, Nov. 2, 2007.

- Gibbs, H. M., "Photonic materials and devices," Karlsruhe School of Optics and Photonics, Karlsruhe, Germany, Nov. 5, 2007; plenary lecture during Karlsruhe Days of Optics and Photonics.
- Gibbs, H. M., "Semiconductor cavity QED," Institute of Optics and Physics Joint Colloquium, University of Rochester, March 17, 2008a.
- Gibbs, H. M., "Semiconductor cavity QED," invited talk 1.14.2 in the Symposium: *Coherent Control of the Fundamental Processes in Optics*, July 1-4, 2008b, part of the 17<sup>th</sup> International Laser Physics Workshops (LPHYS'08), Trondheim, Norway.
- Hendrickson, J., B. C. Richards, J. Sweet, S. Mosor, C. Christenson, D. Lam, G. Khitrova, H. M. Gibbs, T. Yoshie, A. Scherer, O. B. Shchekin, and D. G. Deppe, "Quantum dot photonic-crystal-slab nanocavities: quality factors and lasing," *Phys. Rev. B* **72**, 193303 (2005a).
- Hendrickson, J., S. Mosor, B. C. Richards, J. Sweet, G. Khitrova, H. M. Gibbs, T. Yoshie, A. Scherer, O. B. Shchekin, and D. G. Deppe, "Scanning a photonic crystal slab nanocavity by condensation of xenon for cavity QED experiments," *Frontiers in Optics 2005 - 89th OSA Annual Meeting*, Tucson, Oct. 16-20, 2005b postdeadline talk PDP-A2.
- Hendrickson, J., S. Mosor, B. C. Richards, J. Sweet, G. Khitrova, H. M. Gibbs, T. Yoshie, A. Scherer, O. B. Shchekin, and D. G. Deppe, "Quantum optics in photonic crystal cavities," PQE-2006, Snowbird, Utah, Jan. 2-6, 2006a. Invited talk by Hendrickson.
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- Khitrova, G., H. M. Gibbs, M. Kira, S. W. Koch, and A. Scherer, "Vacuum Rabi splitting in semiconductors," *Nature Physics* **2**, 81-90 (2006).
- Khitrova, G., H. M. Gibbs, and A. Scherer, "InAs quantum dots in photonic crystal nanocavities," invited talk at FOPS 2007, July 22-27, Big Sky, Montana.
- Khitrova, G., "Single quantum dot spectroscopy in a cavity," invited talk CThS3, CLEO2008, San Jose, May 8, 2008a.
- Khitrova, G., "Semiconductor cavity QED; radiatively coupled Fibonacci quantum wells," invited talk QO II-3, Ninth International Workshop on *Nonlinear Optics and Excitation Kinetics in Semiconductors*, Klink, Germany, May 25-29, 2008b.
- Khitrova, G., "Radiatively coupled Fibonacci quantum wells," invited talk 1.14.3 in the Symposium: *Coherent Control of the Fundamental Processes in Optics*, July 1-4, 2008c, part of the 17<sup>th</sup> International Laser Physics Workshops (LPHYS'08), Trondheim, Norway.

- Mosor, S., J. Hendrickson, B. C. Richards, J. Sweet, G. Khitrova, H. M. Gibbs, T. Yoshie, A. Scherer, O. B. Shchekin, and D. G. Deppe, "Scanning a photonic crystal slab nanocavity by condensation of xenon," *Appl. Phys. Lett.* **87**, 141105 (2005).
- Richards, B. C., J. Hendrickson, J. Sweet, C. Christenson, M. Pajor, D. Lam, G. Khitrova, H. M. Gibbs, A. Scherer, and U. Khankhoje, "Emission characteristics of quantum dots in photonic crystal nanocavities," Poster #13 at FOPS 2007, July 22-27, Big Sky, Montana.
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- Soukoulis, C. M., S. Linden, and M. Wegener, "Negative refractive index at optical wavelengths," *Science* **315**, 47 (2007).
- Sünner, T., C. Schneider, M. Strauss, A. Huggenberger, D. Wiener, S. Höfling, M. Kamp, and A. Forchel, "Scalable fabrication of optical resonators with embedded site-controlled quantum dots," *Opt. Lett.* **33**, 1759 (2008).
- Sweet, J., C. Christenson, B. C. Richards, J. Hendrickson, G. Khitrova, H. M. Gibbs, A. Scherer, and Srinivas Manne, "Photoluminescence lifetimes of quantum dot ensembles," Poster #18 at FOPS 2007, July 22-27, Big Sky, Montana.
- Wegener, M., J. L. G. Pomar, N. Meizner, M. Ruther, and S. Linden, "Toy model for metamaterials incorporating gain," summary for 2008 Rochester topical meeting on metamaterials.
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- Zheludev, N. I., S. L. Prosvirnin, N. Papasimakis, and V. A. Fedotov, "Coherent metamaterials and the lasing spaser," *Nature Photonics* **2**, 351-354 (2008).

### Personnel

Graduate students supported by this grant: Sorin Mosor (graduated with Ph.D. in December 2005), Mike Pajor (graduated with M.S. in May 2005), and Josh Hendrickson. Professor: Galina Khitrova; also associated with the research was Professor Hyatt Gibbs.

### Publications

- S. Mosor, J. Hendrickson, B. C. Richards, J. Sweet, G. Khitrova, H. M. Gibbs, T. Yoshie, A. Scherer, O. B. Shchekin, and D. G. Deppe, "Scanning a photonic crystal slab nanocavity by condensation of xenon," *Appl. Phys. Lett.* **87**, 141105 (2005).
- J. Hendrickson, B. C. Richards, J. Sweet, S. Mosor, C. Christenson, D. Lam, G. Khitrova, H. M. Gibbs, T. Yoshie, A. Scherer, O. B. Shchekin, and D. G. Deppe, "Quantum dot photonic-crystal-slab nanocavities: quality factors and lasing," *Phys. Rev. B* **72**, 193303 (2005).
- G. Khitrova, H. M. Gibbs, M. Kira, S. W. Koch, and A. Scherer, "Vacuum Rabi splitting in semiconductors," *Nature Physics* **2**, 81-90 (2006).
- Koch, S. W., M. Kira, G. Khitrova, and H. M. Gibbs, "Semiconductor excitons in new light," *Nature Materials* **5**, 523-531 (2006).

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- G. Khitrova and H. M. Gibbs, "Join the dots," *Nature* **451**, 256 (2008).
- J. Hendrickson, B. C. Richards, J. Sweet, G. Khitrova, A. N. Poddubny, E. L. Ivchenko, M. Wegener, and H. M. Gibbs, "Exciton polaritons in Fibonacci quasicrystals," *Opt. Express*, to be published.

### **Interactions/Transitions**

Publications, talks at various meetings, and discussions with multiple visitors constitute the technology transfer for this project.

### **Patents**

None.

### **Honors/Awards**

PI Galina Khitrova was elected a Fellow of the Optical Society of America in 2007.